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Warszawa, April 9, 2004

**Re: the first Written Opinion mailed on February 11, 2004  
issued with respect to International Patent Application No.  
PCT/PL03/00040 in the name of Ammono Sp. z o.o. et al.  
Due date: April 11, 2003**

Dear Sirs,

This is the reply to the first Written Opinion under PCT Rule 66 as mailed on February 11, 2004.

1. The Applicants have decided to request detail substantive examination of the case and thus such an examination is hereby respectfully requested.

It is further requested that the second Written Opinion be issued, taking into consideration the arguments presented below in favor of the novelty of the present invention as published under WO 2004/003261A1 over FR 2 796 657 A mentioned in the International Search Report as the citation destroying novelty of claims 1, 2, 4-6 and 21-25 as filed (and as published).

2. After thorough consideration of the disclosure contained in the publication FR 2 796 657 A the Applicants are of the opinion that following points should be discusses in order to demonstrate novelty of the present invention over the prior art known form the a.m. French patent application.

The cited publication is directed to a solvothermal crystallogenesi process claimed to allow obtaining mono-crystals of gallium nitride and other nitrides of Group XIII elements. The process involves use of nitrogenous solvent in supercritical state, as well as mother substance and also of crystallization seeds composed of segments of mono-crystalline or polycrystalline material (page 6 last paragraph). It is foreseen that in the known process additives increasing solvent's nitriding power may be used. Among other such additives  $\text{NaN}_3$  is mentioned.

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However, according to FR 2 796 657 A the best results are obtained when additive produces – upon its decomposition, a hydrogenated atmosphere, which is not the case when  $\text{NaN}_3$  is used.

Furthermore, it is mentioned in FR 2 796 657 A that decomposition of additive used should result in subproducts easy to be separated from desired gallium nitride. It is specifically pointed out that “when  $\text{NaN}_3$  is used as an additive, elimination of subproducts containing sodium is difficult and even harmful for GaN microcrystallites, due to the alteration of their surface.” (page 4, lines 30-31 through page 5, line 1)

Although the publication FR 2 796 657 A fails to confirm the information provided therein with any experimental data such as features of the obtained mono-crystalline gallium-containing nitrides, it is clear that when following the teaching contained in FR 2 796 657 A - use of azide mineralizers would be rather avoided.

The experimental research and studies conducted thus far by the present Applicants were concluded by a novel finding that the solubility of gallium-containing nitride decreases with temperature and increases with pressure within the reported temperature and pressure ranges in a supercritical ammonia solution comprising alkali metal ions and soluble forms of gallium.

On the basis of this fundamental finding a controlled selective growth of bulk mono-crystals of gallium-containing nitride, gallium nitride in particular, on crystallization seeds could be designed and carried out in pressurized reactors in the environment of supercritical ammonia solution. In a number of experiments the main problems related to avoidance of both spontaneous crystallization and dissolution of crystallization seeds, and also to ensuring the sufficient rate of dissolution of feedstock, to maintain the appropriate gallium concentration gradient and effective chemical transport between feedstock and seeds had to be identified and solved. Actually the Applicants have successfully solved all the a.m. problems.

According to the present invention new benefits resulting from the use of the specific azide mineralizers are explored. The present application is based on usage of azide mineralizer and it is thus different from previously filed applications Nos. PCT/IB02/04185 and PCT/PL02/00077 (in the name of the same Applicants), which were already examined by the European Patent Office as the International Preliminary Examination Authority under PCT in the last months. On the filing date of the present application No. PCT/PL03/00040 the two a.m. earlier applications were not published as yet.

In order to distinguish further the present invention from the cited French patent application No. FR 2 796 657 A, in which none of the above specified technical problems have been identified or solved, it should be noted that the prior art solvothermal method is a so-called “paper invention” drawn up in full analogy to a hydrothermal method for obtaining mono-crystals of quartz, i.e. it is a process

designed on the assumption of a positive temperature coefficient of solubility of nitrides of Group XIII elements. The arrangement of the mother substance and crystallization seeds presented in the figure accompanying the specification and claims of FR 2 796 657 A proves that the authors of the publication were not aware of the negative temperature coefficient of solubility of GaN and other nitrides of Group XIII elements.

Therefore, apart from the mineralizer, the present invention differs from the prior art known from FR 2 796 657 A due to a difference in arrangement of the feedstock and seeds, optimal for re-crystallizing GaN in nitrogen-containing solvents comprising alkali metals. Thanks to the finding that gallium containing nitride exhibits a negative temperature coefficient of solubility it was possible for the present Applicants to propose an effective method, allowing using the benefits of convection and enabling efficient re-crystallization.

Thus, according to the present invention a selected azide mineralizer (novelty of claims 26-30 has been confirmed in the International Search Report) is used in a process which is distinctively different form the method taught by FR 2 796 657 A.

Only after having carefully selected the dissolution-crystallization process parameters on the basis of the experimentally proven negative temperature coefficient of solubility of the subject nitrides, it has now turned out possible to produce - in a reproducible way - big size bulk mono-crystals of gallium-containing nitrides having the properties required for use as substrates for epitaxy and for manufacturing nitride semiconductor laser structures thereon. Obtaining such crystals was thus far impossible.

Improvement offered by the present invention is advantageous for many reasons specified in the description of the present invention. In short, according to the present invention, not only excellent quality of the product is obtained but also handling of the starting materials and the process itself is much easier.

Actually, the French patent application No. FR 2 796 657 A fails to teach how to obtain satisfactory mono-crystals of gallium-containing nitrides and fails to present any figures characterizing the crystals obtained. The fact that FR 2 796 657 A teaches use of polycrystalline crystallization seeds proves that neither the quality and crystallographic orientation of seeds has been recognized to be crucial factors for producing good quality bulk mono-crystals suitable for use as substrates for epitaxy and manufacturing semiconductor nitride laser structures nor the mono-crystals as covered by the present claim 21 could be and have been obtained according to FR 2 796 657 A.

In view of the above the novelty of the present invention as defined in the claims 1, 2 4-6 and 21-15 over FR 2 796 657 A should thus be recognized and confirmed.

Furthermore, in view of the known facts related to the world-wide competition

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in the attempts of various industrial and scientific centers to develop "blue laser" devices, the fact that on the basis of the French patent application No. FR 2 796 657 A no commercial product has been thus far developed is a crucial proof that the process disclosed in the citation could not be successfully used in practice and that the crystals obtained by the process fail to meet specifications required for use in opto-electronics.

Accordingly, the disclosure of the French application No. FR 2 796 657 A cannot destroy the novelty of the indicated part of the present invention.

3. The Applicants believe that in view of the above explanations the claims presently on file define a patentable invention, i.e. novel, industrially applicable technical solution involving an inventive step, so that a positive International Preliminary Examination Report may be expected and thus issuance of the positive International Preliminary Examination Report is respectfully solicited.

In case the claims cannot be accepted yet, an issuance of a second Written Opinion pursuant to Rule 66 PCT is respectfully requested.

In anticipation of favorable result of the examination, I remain

Yours faithfully,

Ewa Malewska,  
Patent Attorney